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PATENT

Re: Application	: Roger A. Grey et al.	**Case No.**	: 01-2623A
Serial No	: 10/770,924	**Unit No**	: 1625
Filed	: 02/03/2004	**Examiner**	: R. K. Covington
For	: Epoxidation Process Using A Mixed Catalyst System		

Enclosed are the following documents related to the above-identified application:

(X) Return Receipt of Postcard
(X) Certificate of Mailing

( ) Assignment for Recordal
( ) Information Disclosure Statement Under 37 C.F.R. §1.97(b)(1), References, and Form PTO-1449
( ) Amendment After Allowance 37 C.F.R. § 1.312
( ) Response to Restriction Requirement
( ) Two-Month Extension of Time Under 37 C.F.R. § 1.136 (fee noted below)
( ) Amendment Under 37 C.F.R. §1.121

( ) Declaration Under 37 C.F.R. § 1.131
( ) Declaration Under 37 C.F.R. § 1.132
( ) Terminal Disclaimer 37 C.F.R. § 1.321(c)
( ) Notice of Appeal
( ) Appeal Brief Under 37 C.F.R. § 1.192(d)
( ) Version with Markings to Show Changes
( ) Issue Fee Transmittal
( ) Certificate Under 37 C.F.R. § 3.73(b)
( ) Request for Reconsideration
(X) Response to Examiner's Notification of Non-Compliant Appeal Brief (37 CFR § 1.192(d))

The fee has been calculated as shown below:

CLAIMS AS AMENDED

	Claims Remaining After Amendment		Highest No. Previously Paid for	Present Extra	Rate	Add'l Fee
Total Claims:		minus	20		x $50	0.00
Ind. Claims:		minus	3		x $200	0.00
Fee for Petition of Extension of time						0.00
		TOTAL FEE DUE			$	0.00

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01-2623A

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of:

Roger A. Grey et al. : Art Unit: 1625

Serial No.: 10/770,924 : Examiner: R. K. Covington

Filed: 2/3/04

For: EPOXIDATION PROCESS USING A MIXED CATALYST SYSTEM

Commissioner for Patents
P.O. Box 1450
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SECOND CORRECTED APPEAL BRIEF UNDER 37 C.F.R. § 1.192(d)

This is a Second Corrected Appeal Brief from the Final Rejection of claims 1-18. This corrected Appeal Brief is filed in response to Examiner's Notification of Non-Compliant Appeal Brief (37 CFR 41.37), mailed on November 15, 2006. Please replace the previous Appeal Brief filed on October 26, 2006 with the following replacement Appeal Brief in accordance with 37 CFR 41.37(d).

I. Real Party in Interest

The real party in interest is Lyondell Chemical Technology, L.P., a subsidiary of Lyondell Chemical Company.

II. Related Appeals and Interferences

There are no other appeals or interferences known to Appellants, their representative, or assignee that will directly affect or be directly affected by or have a bearing on the Board's decision in this appeal.

III. Status of the Claims

Claims 1-18 are pending in this application. Claims 1-18 have been rejected and are the subject of this appeal.

IV. Status of Amendments

There are no amendments filed subsequent to final rejection. Claims 1-18 on appeal are the originally filed claims. Claims 19-20 were cancelled in a prior response.

V. Summary of Claimed Subject Matter

Appellants' claim 1 claims a process for producing an epoxide comprising reacting an olefin, hydrogen and oxygen in the presence of a catalyst mixture comprising a palladium-containing titanium zeolite and a palladium-free titanium zeolite.

The catalyst mixture comprises a palladium-containing titanium zeolite (discussed at p. 3, l. 4 to p. 4, l. 17). The catalyst mixture also comprises a palladium-free titanium zeolite (discussed at p. 4, l. 18-22). The epoxidation process is discussed in detail at page 5, l. 3 to page 7, l. 14. The olefin is discussed at page 5, l. 4-13. Oxygen and hydrogen are discussed at page 5, l. 14-16.

Appellants claim 14 claims a process comprising reacting propylene, hydrogen and oxygen in a solvent in the presence of a catalyst mixture comprising a palladium-containing titanium silicalite and palladium-free TS-1, wherein the palladium-containing titanium silicalite comprises palladium and a titanium silicalite.

The epoxidation process is discussed in detail at page 5, l. 3 to page 7, l. 14. Propylene is discussed as an especially useful olefin at page 5, l. 11-13. Oxygen and hydrogen are discussed at page 5, l. 14-16. The solvent is discussed at page 6, l. 21-27. The catalyst mixture comprises a palladium-containing titanium zeolite (discussed at p. 3, l. 4 to p. 4, l. 17). The catalyst mixture also comprises a palladium-free titanium zeolite (TS-1) (discussed at p. 4, l. 18-22). TS-1 is discussed as a particularly preferred titanium zeolite at page 3, l. 15-20.

VI. Grounds of Rejection to be Reviewed on Appeal

A. Are claims 1-18 obvious under 35 U.S.C. § 103(a) as unpatentable over Jones (U.S. Pat. No. 6,307,073) in view of Sato et al. (JP 4-352771)?

B. Are claims 1-18 obvious under 35 U.S.C. § 103(a) as unpatentable over Grey et al. (U.S. Pat. No. 6,498,259) in view of Bowman et al. (WO 98/00413)?

VII. Argument

A. Claims 1-18 are Nonobvious over Jones in view of Sato

1. Jones (U.S. Pat. No. 6,307,073) and Sato et al. (JP 4-352771)

Jones teaches an epoxidation process comprising reacting an olefin, hydrogen and oxygen in an oxygenated solvent in the presence of a catalyst mixture. The catalyst mixture of Jones comprises (1) a titanium zeolite; and (2) a supported catalyst comprising gold and a support, wherein the support is an inorganic oxide containing titanium or zirconium (see Claim 1). The support is preferably titania, zirconia, or titania-silica (see Claim 5).

Sato (JP 4-352771) discloses the production of propylene oxide from the reaction of propylene, oxygen, and hydrogen using a catalyst comprising a Group VIII metal such as palladium and a crystalline titanosilicate (see page 2 of translation, Claim 1). Sato teaches that "the Group VIII metal may be supported on the crystalline titanosilicate, or first supported on silica, alumina, activated carbon or the like and then physically mixed with the titanosilicate" (see p. 4, para. 11). Thus, Sato teaches one of two kinds of catalysts for the epoxidation of propylene with oxygen and hydrogen:

(1) Pd/titanosilicate (i.e., palladium supported on a titanosilicate); or

(2) A mixture of (a) a titanosilicate; and (b) a Pd/support (i.e., palladium supported on a support such as silica).

2. The PTO Position: Appellants' Claimed Process is Obvious over Jones in view of Sato

The Examiner's main argument is located in the Office Action dated February 23, 2005. The Examiner states that "Jones differs from the claimed invention in that it does not explicitly include a palladium component .... However, Jones does disclose that [it] is well known for use in these types of processes in order to promote the in-situ

formation of the oxidizing agent. See Col. 1, l. 41-46." Examiner concludes that because Sato teaches a catalyst comprising palladium and a crystalline titanosilicate, "it would have been obvious to one of ordinary skill in the art to combine the teaches of Jones and JP 4-352771 in order to obtain their cumulative effects."

3. The Cited Prior Art Does Not Suggest to One of Ordinary Skill in the Art That They Should Make Appellants' Claimed Process

"A proper analysis under § 103 requires, inter alia, consideration of two factors: (1) whether the prior art would have suggested to those of ordinary skill in the art that they should make the claimed composition or device, or carry out the claimed process; and (2) whether the prior art would also have revealed that in so making or carrying out, those of ordinary skill would have a reasonable expectation of success" (emphasis added). *In re Vaeck*, 947 F.2d 488 (Fed. Cir. 1991). The cited patents neither suggest Appellants' claimed invention, nor would give one of ordinary skill a reasonable expectation that the claimed process would be successful.

Appellants' current application claims an epoxidation process comprising reacting an olefin, hydrogen and oxygen in an oxygenated solvent in the presence of a catalyst mixture. The catalyst mixture comprises (1) a palladium-containing titanium zeolite; and (2) and a palladium-free titanium zeolite.

Examiner has cited Jones as the basis for the § 103(a) rejection. Like Appellants' claimed invention, Jones teaches an epoxidation process comprising reacting olefin, hydrogen and oxygen to form an epoxide. Jones also teaches the use of a catalyst mixture, but the catalyst mixture is different from the mixture required in the current claimed invention (see Table 1 below).

Table 1: Comparison of Jones and Appellants' Catalyst Mixtures

	Appellants' Catalyst Mixture	**Jones**
Component #1	Pd/titanosilicate	Au/TiO_2 (ZrO_2, TiO_2-SiO_2)
Component #2	Titanosilicate (Pd-free)	Titanosilicate

As seen in the above Table, Jones teaches the same second component as required by the present invention - a palladium-free titanosilicate. However, Jones teaches a completely different first component. Jones discloses a Au/TiO_2 (demonstrated in the Examples) as opposed to the Pd/titanosilicate required by Appellants' claimed invention.

Although Examiner notes the differences between Jones and the present invention, Examiner fills the gap with the teachings of Sato. Sato discloses a Pd/titanosilicate catalyst. However, Examiner has failed to describe where the cited prior art would have suggested the combination.

First, one of ordinary skill in the art would have known that a titanium zeolite, such as a titanosilicate, is useful in the epoxidation of olefins with hydrogen peroxide. See Jones at Col. 1, l. 24-27 or Sato at p. 2. para. 3. Second, one of ordinary skill in the art would also have known that palladium promotes the formation of the in situ oxidizing agent, i.e., hydrogen peroxide. See Jones at Col. 1, l. 41-46. Thus, one of ordinary skill in the art would be aware that the epoxidation of olefin with hydrogen and oxygen is performed in the presence of a catalyst that contains both palladium and titanosilicate. The epoxidation reaction is actually two separate, simultaneous reactions. The two separate, simultaneous reactions are catalyzed by the two individual parts of the catalyst system. The two separate parts of the catalyst system are: (1) palladium (or other noble metal) – to promote the formation of in situ hydrogen peroxide from hydrogen and oxygen; and (2) titanosilicate – as a catalyst for the reaction of hydrogen peroxide and olefin to form epoxide. See Figure 1 below.

Figure 1: One Pot/Two Step Olefin + Hydrogen + Oxygen Process

Step 1. Palladium: $H_2 + O_2 \rightarrow H_2O_2$

Step 2. Titanium Zeolite (e.g., titanosilicate): Olefin + $H_2O_2 \rightarrow$ Epoxide

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In fact, Sato (published 12/7/1992) was the first to teach that olefin epoxidation with hydrogen and oxygen could be performed with a catalyst that contained both a titanosilicate and palladium. In the first embodiment of Sato, the two components may be present on the same catalyst particle, i.e., a Pd/titanosilicate. In the second embodiment of Sato, the two components may also be present as a catalyst mixture of two separate particles, i.e., titanosilicate + Pd/silica (alumina, carbon).

Like the second embodiment of Sato, Jones teaches a catalyst mixture of two separate particles. However, Jones does not claim the use of palladium, but rather teaches the use of gold as the noble metal. Specifically, Jones is directed at the use of catalyst mixtures of titanium zeolite and a gold-supported catalyst. Jones, acknowledging Sato and the teachings of the prior art, states that "this reaction is performed in the absence of palladium, which has typically been required in previous liquid-phase direct oxidation processes." In the case of Jones, the Au/TiO_2 is useful to produce hydrogen peroxide in situ, while the titanium zeolite converts olefin and the in situ hydrogen peroxide to epoxide.

Thus, one of ordinary skill in the art might arguably look to Jones to suggest replacing Au/TiO_2 (the in situ hydrogen peroxide forming agent) with any component that is known to produce in situ hydrogen peroxide from the reaction of hydrogen and oxygen. However, one of ordinary skill in the art would have no reason to replace the Au/TiO_2 of Jones with the Pd/titanosilicate of Sato. Since Sato teaches that Pd/titanosilicate is an alternative embodiment to the catalyst mixture embodiment, one may be motivated to replace both the titanosilicate and the Au/TiO_2 of Jones with the Pd/titanosilicate of Sato. However, there is no motivation to combine a Pd/titanosilicate with a palladium-free titanosilicate. As discussed above, Pd/titanosilicate performs both steps of the direct epoxidation reaction: (1) it forms in situ H_2O_2; but simultaneously (2) reacts the H_2O_2 with olefin to produce epoxide.

Thus, one of ordinary skill would not be motivated to combine a palladium-containing titanium zeolite with a palladium-free titanium zeolite because the palladium-

containing titanium zeolite (by itself) is known to be a catalyst useful for olefin epoxidation with hydrogen and oxygen.

In addition, Appellants have demonstrated that although palladium-containing titanium zeolite produces epoxide in the reaction of olefin, hydrogen and oxygen (as was known), the addition of a palladium-free titanium zeolite to the palladium-containing titanium zeolite surprisingly results in enhanced productivity per amount of palladium. Examples 3 and 4 of the current application demonstrate 17-36% higher palladium productivity which as described can result in economic savings by requiring the processing of less TS-1 in palladium incorporation.

B. Claims 1-18 are Nonobvious over Grey in view of Bowman

1. Grey et al. (U.S. Pat. No. 6,498,259) and Bowman et al. (WO 98/00413)

Grey discloses a process for producing an epoxide comprising reacting an olefin, hydrogen and oxygen in a solvent in the presence of a catalyst mixture, wherein the solvent contains a buffer and the catalyst mixture comprises a titanium zeolite and a noble metal catalyst (see Claim 1). The noble metal catalyst is preferably taught to be Pd (or Au) on a support such as carbon, titania, zirconia, niobium oxides, silica, alumina, silica-alumina, tantalum oxides, molybdenum oxides, tungsten oxides, titania-silica, zirconia-silica, or niobia-silica (see Claims 5 and 7).

Bowman teaches a process of preparing an epoxide comprising contacting an olefin with oxygen in the presence of hydrogen and an optional diluent, in the presence of a catalyst comprising gold on a titanosilicate (see Claim 1).

2. The PTO Position: Appellants' Claimed Process is Obvious over Grey in view of Bowman

The Examiner's main argument is located in the Office Action dated February 23, 2005. The Examiner states that "while Grey et al discloses using titanium without catalyst with titanium containing palladium catalyst, it may be argued that a palladium free titanium catalyst is not disclosed.... However, Bowan et al discloses that titanium catalyst compositions falling within the scope of Grey et al are considered palladium

free when the amount of palladium is less than 0.01 in analogous process.... In view of the art as a whole it would have been obvious to one of ordinary skill in the art to modify the process of Grey et al to incorporate the teachings of Bowan et al in order to obtain the cumulative effect of the mixed catalyst system."

3. The Cited Prior Art Does Not Suggest to One of Ordinary Skill in the Art That They Should Make Appellants' Claimed Process

Appellants' current application claims an epoxidation process comprising reacting an olefin, hydrogen and oxygen in an oxygenated solvent in the presence of a catalyst mixture. The catalyst mixture comprises (1) a palladium-containing titanium zeolite; and (2) and a palladium-free titanium zeolite.

Examiner has cited Grey as the basis for the § 103(a) rejection. Like Appellants' claimed invention, Grey teaches an epoxidation process comprising reacting olefin, hydrogen and oxygen to form an epoxide. Grey also teaches the use of a catalyst mixture, but the catalyst mixture is different from the mixture required in the current claimed invention (see Table 2 below).

Table 2: Comparison of Grey and Appellants' Catalyst Mixtures

	Appellants' Catalyst Mixture	Grey
Component #1	Pd/titanosilicate	Pd or Au/C (TiO_2, ZrO_2, SiO_2...)
Component #2	Titanosilicate (Pd-free)	Titanosilicate

As seen in the above Table, Grey teaches the same second component as required by the present invention - a palladium-free titanosilicate. However, Grey teaches a completely different first component. Jones discloses a Pd/C (or Nb_2O_5) or Au/TiO_2 (demonstrated in the Examples) as opposed to the Pd/titanosilicate required by Appellants' claimed invention.

Although Examiner notes the differences between Grey and the present invention, Examiner fills the gap with the teachings of Bowman which teaches a Au/titanosilicate catalyst. However, Examiner has failed to describe where the cited prior art would have suggested the combination.

First, as described above, it was well known that titanosilicate catalyzes the epoxidation of olefins with hydrogen peroxide. See Grey at Col. 1, l. 27-30. Second, as described above, it was well known that noble metals (such as palladium and gold) promote the formation of the in situ oxidizing agent, i.e., hydrogen peroxide. Thus, one of ordinary skill in the art would be aware that the epoxidation of olefin with hydrogen and oxygen is performed in the presence of a catalyst that contains both a noble metal and titanosilicate. Two separate, simultaneous reactions are catalyzed by the two individual parts of the catalyst system: (1) a noble metal – to promote the formation of in situ hydrogen peroxide from hydrogen and oxygen; and (2) titanosilicate – as a catalyst for the reaction of hydrogen peroxide and olefin to form epoxide. As discussed above, these two individual parts may be present on the same catalyst particle, i.e., a Au/titanosilicate, as disclosed in Bowman. In addition, the two components may also be present as two separate particles, i.e., titanosilicate + Pd/silica (alumina, carbon), as disclosed in Grey.

Thus, one of ordinary skill in the art might arguably look to Grey (as with Jones) to suggest replacing the Pd/C (silica,...) (the in situ hydrogen peroxide forming agent) with any component that is known to produce in situ hydrogen peroxide from the reaction of hydrogen and oxygen. However, one of ordinary skill in the art would have no reason to replace Grey's Pd/C with Bowman's Au/titanosilicate. Since Bowman teaches that Au/titanosilicate catalyzes the entire reaction by itself, one may be motivated to replace both the titanosilicate and the Pd/C of Grey with the Au/titanosilicate of Bowman. However, since Bowman's Au/titanosilicate performs both steps of the direct epoxidation reaction [(1) it forms in situ H_2O_2; but simultaneously (2) reacts the H_2O_2 with olefin to produce epoxide], there is no motivation to combine a Au/titanosilicate with a palladium-free titanosilicate.

Thus, one of ordinary skill would not be motivated to combine a gold-containing titanium zeolite (let alone a palladium-containing titanium zeolite) with a palladium-free titanium zeolite because the gold-containing titanium zeolite (by itself) is known to be a catalyst useful for olefin epoxidation with hydrogen and oxygen.

In addition, neither Grey nor Bowman suggests Appellants' surprising results of enhanced productivity per amount of palladium.

The § 103 analysis thus fails under both factors described above. First, the combination of Grey and Bowman would not suggest to one of ordinary skill in the art that they should make Appellants' claimed process. Second, Grey and Bowman do not reveal to those of ordinary skill would have a reasonable expectation of success that in so carrying out Appellants' claimed process one would get enhanced productivity per amount of palladium as demonstrated in the current application.

C. Summary

In sum, Jones and Sato or Grey and Bowman fail to teach or suggest a process. Because the cited prior art does not suggest Appellants' claimed process, the claimed process meets the patentability requirements of Section 103(a).

Because the differences between the claimed process and those disclosed by Jones and Sato or Grey and Bowman would not have been obvious to a sk0illed person in this field, the Board should reverse the Section 103 rejection.

VIII. Conclusion

Appellants respectfully ask the Board of Patent Appeals and Interferences to reverse the Examiner's rejections of claims 1-18 under 35 U.S.C. § 103(a).

I hereby certify that this correspondence is being deposited with the United States Postal Service as first-class mail, with sufficient postage, in an envelope addressed to: Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450 on December 15, 2006.

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Respectfully submitted,

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Claims Appendix

1. (original) A process for producing an epoxide comprising reacting an olefin, hydrogen and oxygen in the presence of a catalyst mixture comprising a palladium-containing titanium zeolite and a palladium-free titanium zeolite.

2. (original) The process of claim **1** wherein the palladium-containing titanium zeolite comprises palladium and a titanium silicalite.

3. (original) The process of claim **2** wherein the titanium silicalite is TS-1.

4. (original) The process of claim **1** wherein the palladium-containing titanium zeolite comprises palladium, a titanium zeolite, and a noble metal selected from the group consisting of platinum, gold, silver, iridium, rhenium, ruthenium, osmium, and mixtures thereof.

5. (original) The process of claim **4** wherein the noble metal is selected from the group consisting of platinum, gold, and mixtures thereof.

6. (original) The process of claim **1** wherein the palladium-containing titanium zeolite comprises from about 0.01 to about 10 weight percent palladium.

7. (original) The process of claim **1** wherein the palladium-free titanium zeolite is a titanium silicalite.

8. (original) The process of claim **1** wherein the palladium-free titanium zeolite is TS-1.

9. (original) The process of claim **1** wherein the olefin is a C_2-C_6 olefin.

10. (original) The process of claim **1** wherein the olefin is propylene.

11. (original) The process of claim **1** wherein reaction of olefin, hydrogen and oxygen is performed in a solvent.

12. (original) The process of claim **11** wherein the solvent is selected from the group consisting of water, C_1-C_4 alcohols, supercritical CO_2, and mixtures thereof.

13. (original) The process of claim **11** wherein the solvent contains a buffer.

14. (original) A process comprising reacting propylene, hydrogen and oxygen in a solvent in the presence of a catalyst mixture comprising a palladium-containing titanium silicalite and palladium-free TS-1, wherein the palladium-containing titanium silicalite comprises palladium and a titanium silicalite.

15. (original) The process of claim **14** wherein the titanium silicalite is TS-1.

16. (original) The process of claim **14** wherein the palladium-containing titanium zeolite further comprises a noble metal selected from the group consisting of platinum, gold, silver, iridium, rhenium, ruthenium, osmium, and mixtures thereof.

17. (original) The process of claim **14** wherein the solvent is selected from the group consisting of water, C_1-C_4 alcohols, supercritical CO_2, and mixtures thereof.

18. (original) The process of claim **14** wherein the solvent contains a buffer.

19. (cancelled)

20. (cancelled)

Evidence Appendix

None

Related Proceedings Appendix

None